



(19)

Europäisches Patentamt

European Patent Office

Office européen des brevets



(11)

**EP 0 842 894 A1**

(12)

**EUROPEAN PATENT APPLICATION**

(43) Date of publication:  
20.05.1998 Bulletin 1998/21

(51) Int. Cl.<sup>6</sup>: **C01B 3/38**

(21) Application number: **97119515.1**

(22) Date of filing: **07.11.1997**

(84) Designated Contracting States:  
**AT BE CH DE DK ES FI FR GB GR IE IT LI LU MC  
NL PT SE**

(71) Applicant: **Haldor Topsoe A/S  
DK-2800 Lyngby (DK)**

(30) Priority: **15.11.1996 US 30847 P**

(72) Inventor: **Primdahl, Ivar Ivarsen  
2400 Copenhagen NV (DK)**

(54) **Process and apparatus for catalytic partial oxidation of a hydrocarbon**

(57) Process and apparatus for the preparation of a hydrogen/carbon monoxide containing synthesis gas by catalytic partial oxidation reaction of a hydrocarbon feed gas by carrying out the process in a number of separated steps. In each step only a small fraction of the stoichiometric amount of oxygen is added to the reaction and the feed gas is passed through a catalytic zone arranged in each step, so that complete conversion of the added amount of oxygen is obtained in each step.

**EP 0 842 894 A1**

## Description

### BACKGROUND OF THE INVENTION

It is known to prepare synthesis gas compositions consisting of hydrogen and carbon monoxide and containing very low content of methane and carbon dioxide by passing over a catalyst of hydrocarbon substrate, usually natural gas or methane mixed with oxygen at preheated temperature in the range of, e.g. 300-400°C.

The processes are typically performed in the presence of a noble metal catalyst, such as rhodium, platinum, palladium, or iridium, or other metals like nickel, cobalt and ruthenium, arranged in a fixed bed reactor. The residence time of the gas in a catalytic bed thereby is very short. Catalytic material can furthermore be in form of a sponge metal, a metal web or supported on ceramic porous bodies, for example.

By the above process, it is possible to obtain a substantial and complete conversion of hydrocarbon with high selectivity for carbon monoxide and hydrogen. In order to obtain high selectivity, the residence time is usually very low and the gas maintained at elevated temperature for the shortest time possible.

Alternatively, selectivity is achieved by keeping the process temperature as low as possible. The main problem with the known process is to avoid ignition of the gas/oxygen mixture prior to contact with the catalyst. In particular, this is problematic when the process is carried out at elevated pressure.

### SUMMARY OF THE INVENTION

This invention provides an improved process for catalytic partial oxidation of a hydrocarbon substrate of the above type. The improvement comprises carrying out the process in a number of separated steps, whereby in each step solely a small fraction of the stoichiometric amount of oxygen is added to the reaction. The gas is passed through a catalytic zone arranged in each step, so that complete conversion of oxygen is obtained in each step. The reaction is performed at substantially the same temperature in all steps by cooling the reaction mixture in each step with a suitable cooling medium during the total process cycle.

### BRIEF DESCRIPTION OF THE DRAWING

The sole figure is a schematic cross-section of a reaction apparatus according to a preferred embodiment of the present invention.

### DESCRIPTION OF THE PREFERRED EMBODIMENT

Referring to the sole figure, a reaction apparatus according to a specific embodiment of the invention is shown schematically in a representative cross-section. The process is performed on a hydrocarbon gas sub-

strate 2 fed to a tubular reactor surrounded by a cooling medium, such as pressurized boiling water 4, contained between reactor wall 6 and jacket 8 provided in the form of concentric tubes.

The reactor is further provided with static mixing zones 10 in the reaction volume. Toward the beginning of each mixing zone, oxygen 12 is provided through perforated rings 14. Catalyst bodies 16 are arranged at predetermined distances from the oxygen supply rings. Product 18 is generated after gas 2 has passed through a predetermined number of catalyst reaction zones.

When carrying out the process of this invention as described above, excessive concentrations of oxygen will not be available in the reactor. As a result, a very high temperature in the reactor is prevented, all the catalyst material will be at a controlled temperature, and the selectivity of the process thereby is improved.

The invention further is useful in partial oxidation of higher hydrocarbons and in oxidative coupling reactions.

Although the present invention has been described in relation to particular embodiments thereof, many other variations and modifications and other uses will become apparent to those skilled in the art. It is preferred, therefore, that the present invention be limited not by the specific disclosure herein, but only by the appended claims.

### Claims

1. A process for the preparation of a hydrogen/carbon monoxide containing synthesis gas by catalytic partial oxidation of a hydrocarbon feed gas with a stoichiometric amount of oxygen, the process comprising a plurality of sequential, separated steps, whereby in each step only a small fraction of the stoichiometric amount of oxygen is added to the hydrocarbon feed gas and the feed gas is then passed through a catalytic zone, so that substantially complete conversion of the added amount of oxygen by reaction with a portion of the feed gas is obtained in each step.
2. The process of claim 1, wherein the oxidation reaction is performed at substantially the same temperature in each of the steps by cooling the reaction mixture in each step with a suitable cooling medium.
3. The process of claim 1, wherein each step includes mixing the added amount of oxygen with the feed gas prior to passage through each catalytic zone.
4. The reactor for use in the preparation of a hydrogen/carbon monoxide containing synthesis gas by catalytic partial oxidation of a hydrocarbon feed gas, the preparation being carried out in a number of sequential, separated steps, whereby in each

step only a small fraction of the stoichiometric amount of oxygen is added to the feed gas and the feed gas is then passed through a catalytic zone, so that substantially complete conversion of the added amount of oxygen is obtained in each step, the apparatus comprising a reactor fed by a hydrocarbon gas, the reactor including a plurality of reaction zones through which the hydrocarbon gas passes sequentially, each reaction zone having an oxygen supply and a catalyst, each reaction zone being provided with a cooling medium.

5. The reactor of claim 4, wherein the cooling medium surrounds the reaction zones.

15

6. The reactor of claim 4, wherein the cooling medium is pressurized boiling water.

7. The reactor of claim 4, wherein the reaction zones are provided with static mixing zones between the oxygen supply and the catalyst body, each catalyst body being arranged at a predetermined distance from the oxygen supply.

20

25

30

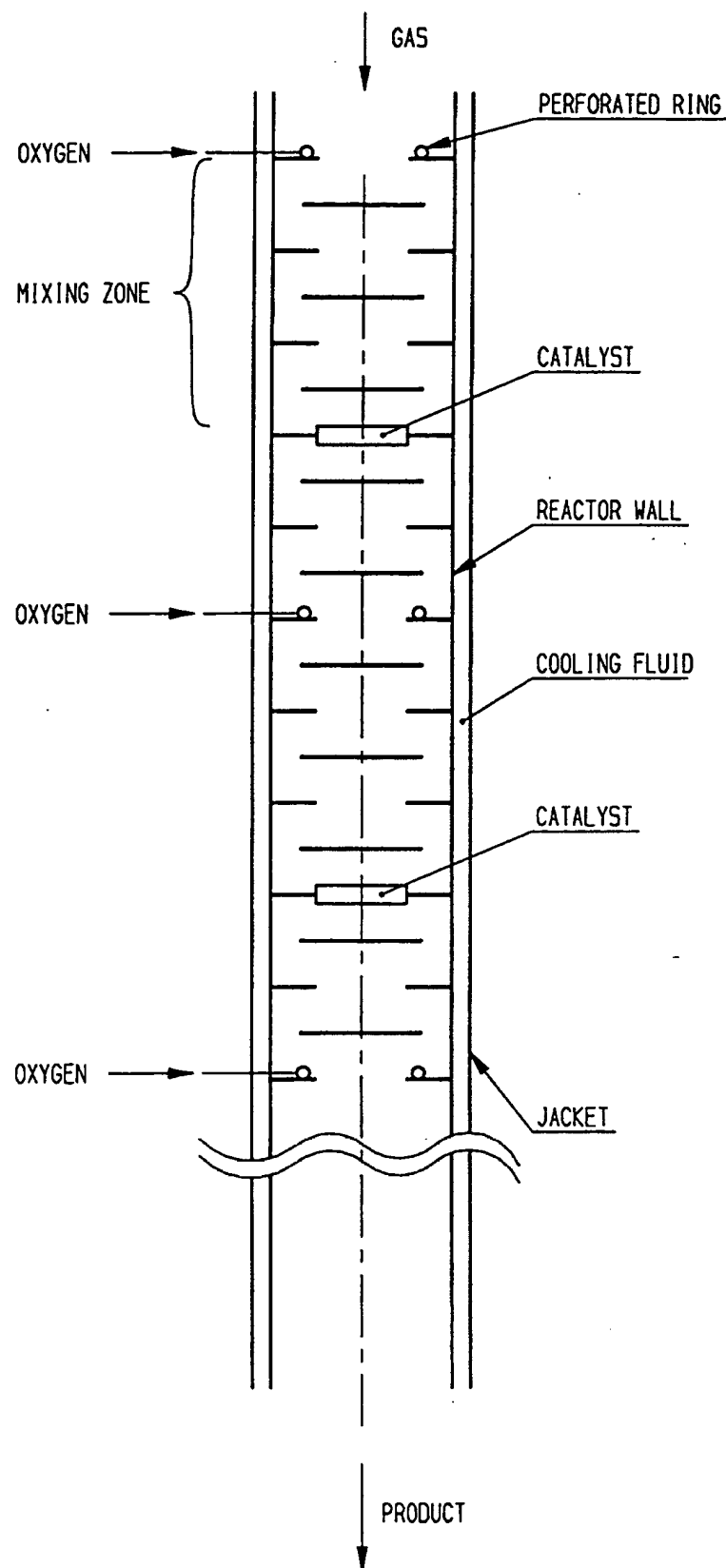
35

40

45

50

55





European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number  
EP 97 11 9515

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	GB 2 274 284 A (SNAM PROGETTI) * page 7, line 16 - page 8, line 14 * * page 9, line 13 - page 11, line 1 * * page 11, line 17 - line 25 * * examples 6-9; tables 1,2 *	1-4	C01B3/38
X	EP 0 330 751 A (UHDE GMBH) * the whole document *	1-4	
A	EP 0 155 867 A (INST FRANCAIS DU PETROL) * page 5, line 5 - page 6, line 31 * * figures 1,2; examples *	1,4	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			C01B
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 20 February 1998	Examiner Van der Poel, W
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			

EPO FORM 1503 03.82 (P04C01)